

# EVIDENCE FOR CHARGE INHOMOGENEITY OF DOPED HOLES IN $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

Does the dopant produce an inhomogeneous hole-state in high  $T_c$  cuprate superconductors? Are the doped holes spatially correlated to Sr dopants in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ? This report addresses these fundamental questions using the x-ray absorption fine structure (XAFS) technique.

The role of dopants in high  $T_c$  superconductors is widely believed to be limited to introducing hole carriers into the  $\text{CuO}_2$  planes of otherwise insulating parent compounds. This simplified assumption is partly driven by the lack of information on the local atomic and electronic structure around dopants. While experimental evidence favoring inhomogeneous charge distributions of the doped holes is still mounting [1], the role that dopants play in determining this inhomogeneous ground state, if any, is still unclear. Since high  $T_c$  superconductors manifest strong carrier-lattice interactions, as evidenced in the presence of *Jahn-Teller* distorted  $\text{CuO}_6$  octahedra, structural techniques can provide information on the spatial distribution of doped charges through the structural response. The x-ray absorption fine structure (XAFS) technique is particularly suited for elucidating such response around dopants as it is element specific. By tuning the x-ray energy to an absorption threshold of a dopant, XAFS determines the *partial* pair correlations between the dopant and its neighbors. This is important since Sr dopants substitute at majority La sites, and techniques that sum over all pair correlations are dominated by those involving La host atoms.

We present evidence that doped holes are spatially correlated to Sr dopants in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . This is seen by the structural response of the Sr-O(2) distance across the  $x \sim 0.06$  insulator-metal transition, while no response is observed for the La-O(2) distance, indicating that the doped charge density is

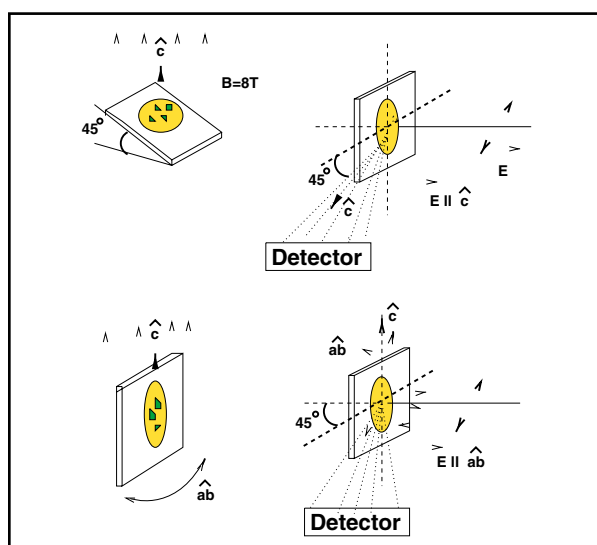


FIG. 1. Special geometry used in the *c*-axis magnetic alignment of powders of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , allowing the measurement of *c* and *ab* polarized fluorescence XAFS in identical geometries; i.e., *E* forming 45° with the surface's normal.

not uniformly distributed and that the dopants play a role in determining the inhomogeneous charge state.

Our experiments are performed on *c*-axis magnetically aligned powders to exploit the angular dependence of XAFS in these anisotropic, layered cuprates. Measuring a subset of the local structure around the absorbing atom allows for a complete determination of its local environment [2]. The main advantages of this method over single crystals are (1) control of sample thickness enabling transmission experiments at absorption edges of concentrated elements (La *K*-edge and Sr *K*-edge for  $x \geq 0.04$ ),

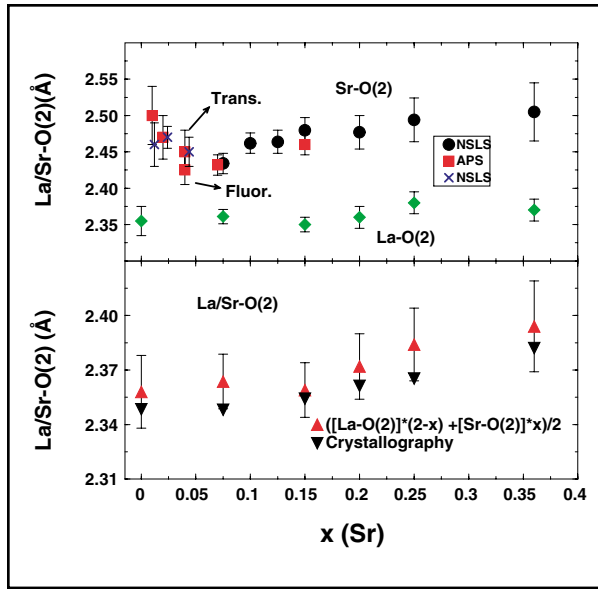


FIG. 2. Local Sr-O(2) and La-O(2) apical distances obtained from  $c$ -axis polarized Sr and La XAFS at their  $K$ -edges at  $T = 10\text{K}$  (top). Sr measurements were done on samples prepared by solid-state reaction (circles) and by precipitation from solution (squares, crosses) and at different synchrotron facilities (APS, NSLS). Results from both transmission and fluorescence measurements are shown for comparison for  $x = 0.04$ . Crosses are displaced in  $x$  for clarity. Bottom: weighted averages of Sr-O(2) and La-O(2) distances as a function of  $x$  and their comparison with the results of crystallography.

and (2) control of  $c$ -axis orientation relative to sample's surface for fluorescence experiments at absorption edges of dilute elements (Sr  $K$ -edge for  $x \leq 0.04$ ). The latter allows measuring  $c$  and  $ab$ -polarized fluorescence XAFS in identical geometries [3,4].

Orientation-dependent spectra were taken by rotating the oriented samples relative to the electric field vector  $E$  of the incident x-rays. Experiments were performed at beamline X11-A of the National Synchrotron Light Source (NSLS, Brookhaven) and at beamline 20-ID-B of the Advanced Photon Source (APS, Argonne).

Figure 2 shows local Sr-O(2) and La-O(2) apical distances determined from  $c$ -axis polarized XAFS at Sr and La  $K$ -edges as a function of Sr content in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  at  $T = 10\text{K}$ . O(2) oxygens are apices of  $\text{CuO}_6$  octahedra, and the La/Sr-O(2) apical bonds nearly coincide with the crystallographic  $c$ -axis [5].

The local Sr-O(2) apical distance is significantly longer than the local La-O(2) distance. Their weighted average,  $[(\text{La-O(2)}) \cdot (2-x) + (\text{Sr-O(2)}) \cdot x] / 2$ , agrees with the values obtained by crystallography, as expected (lower panel of Fig. 1). A longer Sr-O(2) local distance is explained by the weaker attraction of a negatively charged O(2) ion to a divalent  $\text{Sr}^{2+}$  ion than to a trivalent  $\text{La}^{3+}$ . The local La-O(2) distance is nearly independent of  $x$ , and therefore the average expansion of the La/Sr-O(2) distance with  $x$  is due to the increased weight of the long Sr-O(2) distance. A more striking observation is the change in slope in the  $x$  dependence of the Sr-O(2) distance at  $x \sim 0.06$ , while no such change is seen in the La-O(2) distance. We recall that in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  an insulator-metal (I-M) transition takes place at  $x \sim 0.06$ . The fact that the Sr-O(2) distance shows a large response to the delocalization of holes at the I-M transition, but the La-O(2) distance does not, is direct evidence that a spatial correlation exists between the doped holes and the dopants that introduced them. This might not be surprising, since, at low Sr concentrations, the dopants' potential is poorly screened, and it is energetically favorable for a doped hole to remain in the vicinity of the Sr. At larger dopant concentrations, screening becomes more efficient but remains poor at very short distances, so the doped holes, although itinerant, are expected to have significant weight in the vicinity of the dopants.

The structural response to the change in localization of the holes also shows that there is a strong local interaction between the doped hole and the lattice at the dopant sites. We believe this is the first direct experimental evidence for a spatial correlation between dopants and doped holes, although a similar conclusion was derived from the interpretation of NQR data by Hammel et al. [6].

Our experiments with  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  provide evidence for a spatial correlation between doped holes and the Sr dopants that introduced them. A strong hole-lattice coupling in the vicinity of the dopants allowed us to observe this correlation through the structural response of the dopants environment as the holes delocalize at the I-M transition.

This indicates that Sr dopants may play a more significant role in determining normal and superconducting properties of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  than typically assumed.

Research done under auspices of DOE Grant No. DE-FG03-98ER45681. D.H. was also partially supported by DOE Contract No. W-31-109-ENG-38.

Principal publication: “XFAS Study of the Low-Temperature Tetragonal Phase of  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ : Disorder, Stripes and  $T_c$  Suppression at  $x = 0.125$ ,” Phys. Rev. B **61**, 7055-7076 (2000).

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